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EXTENDED CO-ORDINATION THEORY OF VALENCY. III.

Valence Bonds in Carbon Compounds.

By RYUTARO TSUCHIDA AND MASAHISA KOBAYASHI.

Introduction.

No single method of quantum mechanical treatments of valences gives a completely accurate representation of the facts, because mathematical difficulties prohibit the strict calculation save for the simplest types of molecules. Thus the value of various methods so far proposed lies chiefly in the formal representation they afford of the bonding process, and therefore any qualitative methods may be justified as well, so long as they deal with less simple compounds and explain the facts to a certain degree.

Chemical valences are usually classified into electrovalence, covalence and co-ordinate covalence, but it is impossible to draw sharp boundary lines among them: it is a question of degree than of nature. In order to remove this somewhat artificial classification of valences, one of the present authors has ventured to extend Werner's co-ordination theory to the simplest ions and molecules by assuming that all the ions and molecules are built up by co-ordinating ions and molecules around central cations. Thus all the compounds, simple and complex, have been systematized and unified under the extended co-ordination theory.¹⁾ As one of the examples of application of this co-ordination method, the configuration of various compounds of typical elements²⁾, especially in detail of carbon compounds³⁾, has been discussed in the preceeding papers. In the present article, the nature of valence-bonds in carbon compounds is dealt with from a quantum mechanical point of view, in the hope of affording a theoretical basis to this co-ordination method.

Since the *L*-shell can accommodate only for eight electrons in four pairs, the co-ordination number for carbon compounds can not exceed four.²⁾ According to the number and the nature of ligands, however, we can distinguish at least six types³⁾ of carbon compounds, which are shown in Table I.

- 1) R. Tsuchida, *J. Chem. Soc. Japan*, 60, 245 (1939).
- 2) R. Tsuchida, *Bull. Chem. Soc. Japan*, 14, 101 (1939).
- 3) R. Tsuchida, *Rev. Phys. Chem. Japan*, 13, 31 (1939).

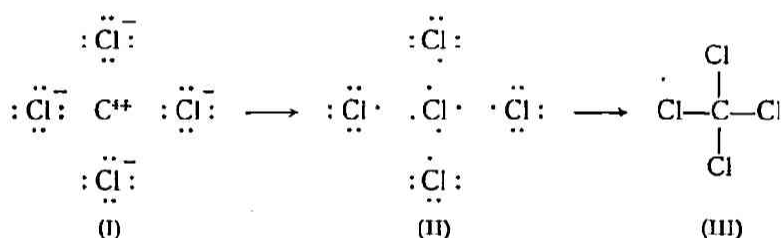
Table I.

Co-ord. number	Number of unsaturated electrons	Configuration	Kinds of bonds	Examples
4	0	tetrahedron	tetrahedral	CCl_4 , CH_4 , etc.
3	0	triangle	trigonal	CO_3^{2-} , $[\text{C}(\text{NH}_2)_3]^+$, RCHO , RCOOH , etc.
3	1	trigonal	aromatic	C_6H_6 , $\text{C}(\text{C}_6\text{H}_5)_3$, etc.
3	1	trigonal	'double'	C_2H_4 , CH_2CHCH_2 , etc.
2	0	linear	digonal	CO_2 , etc.
2	2	digonal	'triple'	C_2H_2 , etc.

The quantizations for these various kinds of bonds will be explained below.

Tetrahedral Quantization.

For example carbon tetrachloride is taken, in which four chlorine anions are assumed to co-ordinate around a quadrivalent carbon cation in the following scheme:



It has already been stated in the previous papers that the four ligands are combined tetrahedrally by the general rule of symmetrical co-ordination, but this configuration may also be derived by Pauling's method⁴⁾ of hybridization. Since the elements to combine are treated as atoms in Pauling's method while ions are dealt with in the present method of co-ordination, the results to be obtained are generally different for both the methods. In special cases, however, they may arrive at the same results. This is the case with tetrahedral carbon compounds. Thus for the state (II) in the above process, the tetrahedral bond eigenfunctions of Pauling can be applied as a whole and therefore it seems unnecessary to repeat the derivation of the functions. It is convenient, however, to refer briefly to the outline of the calculation, for the purpose of showing clearly the assumptions and approximations involved in the method of hybridization and also of facilitating the calculation of the other quantizations to be mentioned below.

Pauling assumed that the eigenfunctions of the carbon atom are hydrogen-

4) L. Pauling, *J. Am. Chem. Soc.*, 53, 1367 (1931).

like ones which may be represented as functions of the form

$$\Psi_{nlm}(r, \theta, \varphi) = R_{nl}(r) \cdot \theta_{lm}(\theta) \cdot \phi_m(\varphi).$$

Then the s and p eigenfunctions are

$$\left. \begin{aligned} \Psi_{200}(r, \theta, \varphi) &= R_{20}(r) \cdot \theta_{00}(\theta) \cdot \phi_0(\varphi) = R_{20}(r) \cdot \frac{1}{\sqrt{2}} \cdot \frac{1}{\sqrt{2\pi}} = R_{20}(r) \cdot \frac{1}{2\sqrt{\pi}}. \\ \Psi_{211}(r, \theta, \varphi) &= R_{21}(r) \cdot \theta_{11}(\theta) \cdot \phi_1(\varphi) = R_{21}(r) \cdot \frac{\sqrt{3}}{2} \sin \theta \cdot \frac{1}{\sqrt{\pi}} \cos \varphi \\ &= R_{21}(r) \cdot \frac{\sqrt{3}}{2\sqrt{\pi}} \sin \theta \cos \varphi. \\ \Psi_{21\bar{1}}(r, \theta, \varphi) &= R_{21}(r) \cdot \theta_{1\bar{1}}(\theta) \cdot \phi_{\bar{1}}(\varphi) = R_{21}(r) \cdot \frac{\sqrt{3}}{2} \sin \theta \cdot \frac{1}{\sqrt{\pi}} \sin \varphi \\ &= R_{21}(r) \cdot \frac{\sqrt{3}}{2\sqrt{\pi}} \sin \theta \sin \varphi. \\ \Psi_{210}(r, \theta, \varphi) &= R_{21}(r) \cdot \theta_{10}(\theta) \cdot \phi_0'(\varphi) = R_{21}(r) \cdot \frac{\sqrt{6}}{2} \cos \theta \cdot \frac{1}{\sqrt{2\pi}} \\ &= R_{21}(r) \cdot \frac{\sqrt{3}}{2\sqrt{\pi}} \cos \theta. \end{aligned} \right\}$$

Since the dependence on r of the s and p eigenfunctions is not greatly different, Pauling assumed that $R_{n0}(r)$ and $R_{nl}(r)$ are effectively the same as far as bond formation is concerned and the problem of determining the bond eigenfunctions reduces to a discussion of the following θ, φ eigenfunctions.

$$\left. \begin{aligned} s &= \frac{1}{2\sqrt{\pi}}. \\ p_x &= \frac{\sqrt{3}}{2\sqrt{\pi}} \sin \theta \cos \varphi. \\ p_y &= \frac{\sqrt{3}}{2\sqrt{\pi}} \sin \theta \sin \varphi. \\ p_z &= \frac{\sqrt{3}}{2\sqrt{\pi}} \cos \theta. \end{aligned} \right\}$$

Normalizing these functions to 4π , Pauling obtained

$$\left. \begin{aligned} s &= 1. \\ p_x &= \sqrt{3} \sin \theta \cos \varphi. \\ p_y &= \sqrt{3} \sin \theta \sin \varphi. \\ p_z &= \sqrt{3} \cos \theta. \end{aligned} \right\} \quad (1)$$

According to Pauling the correct zeroth order bond eigenfunctions are those orthogonal and normalized linear aggregates of both the s and p eigenfunctions as follows:

$$\left. \begin{aligned} \psi_1 &= a_1 s + b_1 p_x + c_1 p_y + d_1 p_z \\ \psi_2 &= a_2 s + b_2 p_x + c_2 p_y + d_2 p_z \\ \psi_3 &= a_3 s + b_3 p_x + c_3 p_y + d_3 p_z \\ \psi_4 &= a_4 s + b_4 p_x + c_4 p_y + d_4 p_z \end{aligned} \right\}$$

The coefficients a_i , b_i , etc. are restricted only by the orthogonality and normalization requirements, i.e., $\int \psi_i^2 d\tau = 1$ and $\int \psi_i \psi_k d\tau = 0$. Thus the tetrahedral eigenfunctions which would give the strongest bonds have been found as

$$\left. \begin{aligned} \psi_1 &= \frac{1}{2} s + \frac{\sqrt{3}}{2} p_x \\ \psi_2 &= \frac{1}{2} s - \frac{1}{2\sqrt{3}} p_x + \frac{\sqrt{2}}{\sqrt{3}} p_z \\ \psi_3 &= \frac{1}{2} s - \frac{1}{2\sqrt{3}} p_x + \frac{1}{\sqrt{2}} p_y - \frac{1}{\sqrt{6}} p_z \\ \psi_4 &= \frac{1}{2} s - \frac{1}{2\sqrt{3}} p_x - \frac{1}{\sqrt{2}} p_y - \frac{1}{\sqrt{6}} p_z \end{aligned} \right\}$$

By a rotation of the atom as a whole, Pauling obtained the following set of four equivalent tetrahedral bond eigenfunctions instead of the above formulae.

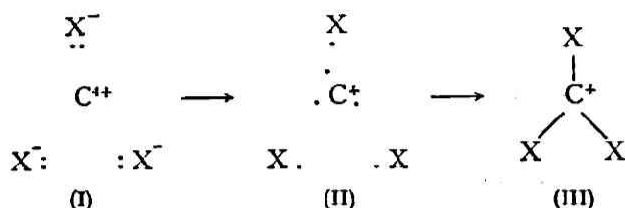
$$\left. \begin{aligned} \psi_{111} &= \frac{1}{2} (s + p_x + p_y + p_z) \\ \psi_{1\bar{1}\bar{1}} &= \frac{1}{2} (s + p_x - p_y - p_z) \\ \psi_{\bar{1}1\bar{1}} &= \frac{1}{2} (s - p_x + p_y - p_z) \\ \psi_{\bar{1}\bar{1}1} &= \frac{1}{2} (s - p_x - p_y + p_z) \end{aligned} \right\} \quad (2)$$

Each of these functions has in the direction of the normal to the tetrahedral face a maximum value of 2.000, considerably larger than 1.732 for the original p function in (1).

It is only for this tetrahedral quantization that the results from Pauling's method of hybridization coincide with those of the present method of co-ordination. In tetrahedral carbon compounds the valency happens to be equal to the co-ordination number, i.e., the number of bonds. There are, however, many cases, in which the valency and the co-ordination number are not equal, and consequently the carbon atom is electrically charged or has one or more unpaired electrons after the ion or molecule is formed. In such cases the tetrahedral eigenfunctions of Pauling are not available.

Trigonal Quantization.

In guanidonium ion three anions NH_2^- are co-ordinated around a quadrivalent carbon cation in the following scheme, NH_2^- being represented by :X^- .



The general rule of symmetrical co-ordination requires that the three ligands are equally distributed around the central ion, and consequently a plane trigonal quantization is required for the three electrons attached to the central ion in the state (II). By similar calculation as above, Pauling⁴⁾ has worked out three equivalent bond eigenfunctions of which the directions of maxima lie in a plane 120° apart.

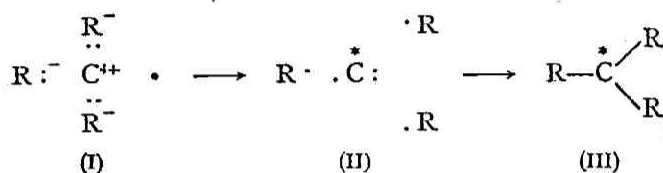
$$\left. \begin{aligned} \psi_1 &= \frac{1}{\sqrt{3}} s + \sqrt{\frac{2}{3}} p_x \\ \psi_2 &= \frac{1}{\sqrt{3}} s - \frac{1}{\sqrt{6}} p_x + \frac{1}{\sqrt{2}} p_y \\ \psi_3 &= \frac{1}{\sqrt{3}} s - \frac{1}{\sqrt{6}} p_x - \frac{1}{\sqrt{2}} p_y \end{aligned} \right\} \quad (3)$$

These functions have a strength of 1.991, only a little less than 2.000 of the tetrahedral bond. Pauling, however, has not placed so much importance in this quantization as in the tetrahedral, but employed the results as a merely auxiliary basis for his discussion of the structure of the carbonate ion. Namely he anticipated for this ion a bond angle larger than $109^\circ 28'$ from the quantization (3). But as the repulsion of the oxygen atom would not be very effective in increasing the bond angle in the neighbourhood of 120° , Pauling expected an equilibrium at a somewhat smaller angle, such as 118° . In order to explain the planar configuration, from the results of X-ray investigations, he assumed a rapid vibration of the carbon atom through the plane of the three oxygen atoms. Such a difficulty seems to have been provoked by the fact that he has persisted in the tetrahedral quantization and slighted the trigonal.

In the present method, on the contrary, the trigonal quantization has nearly the same importance as the tetrahedral. Namely the trigonal quantization (3) is directly applied to the state (II) in which the center is a univalent carbon cation

Aromatic Quantization.

In the present method of co-ordination the center of carbon compounds is consistently the quadrivalent carbon cation. When four ligands are co-ordinated, the tetrahedral configuration results and three ligands give rise to the planar trigonal configuration. Now we can expect a quantization which may be regarded as intermediate between the two. This is the case when one of the ligands is an electron.



In such a case, the three ligands are expected to co-ordinate trigonally by the rule of symmetrical co-ordination and the electron as ligand will remain in the central carbon atom unpaired but not without interaction with other ligands. Such an electron is denoted by an asterisk as above.

The three electrons of the as many ligands in the state (II) must be equivalent and, therefore, be expressed by the trigonal functions (4). The eigenfunction for the lone electron must be certainly normalized and also orthogonal to the former three. Such an eigenfunction can be easily determined. Thus we have

$$\psi_{III} = \frac{1}{\sqrt{3}} (p_x + p_y + p_z). \quad (5)$$

Since this combination of the quantizations (4) and (5) is characteristic of aromatic compounds, the present authors would like to call the following set of eigenfunctions as the aromatic quantization.

$$\left. \begin{aligned} \psi_{I\bar{0}0} &= \frac{1}{\sqrt{3}} (s + p_x - p_y), \\ \psi_{0I\bar{1}} &= \frac{1}{\sqrt{3}} (s + p_y - p_z), \\ \psi_{\bar{1}0I} &= \frac{1}{\sqrt{3}} (s + p_z - p_x), \\ \psi_{III} &= \frac{1}{\sqrt{3}} (p_x + p_y + p_z), \end{aligned} \right\} \quad (6)$$

The former three functions have been explained in the preceding section and we must now examine the fourth. For this purpose, the co-ordinate system

(x, y, z) is transformed into the other (x', y', z') as in Fig. 3. This transformation is carried out as follows. 1) First the system (x, y, z) is rotated for an angle -45° around the Z -axis, i.e., $(x, y, z) \rightarrow (x'', y'', z)$. 2) Next it is rotated for an angle $-\tan^{-1}\sqrt{2}$ around the x'' -axis, i.e., $(x'', y'', z) \rightarrow (x'', y''', z')$. 3) Finally it is rotated through 45° around the z' -axis, i.e., $(x'', y''', z') \rightarrow (x', y', z')$. Let the angles of rotation be α, β and γ in the order of the transformation, then by the formulæ of Euler

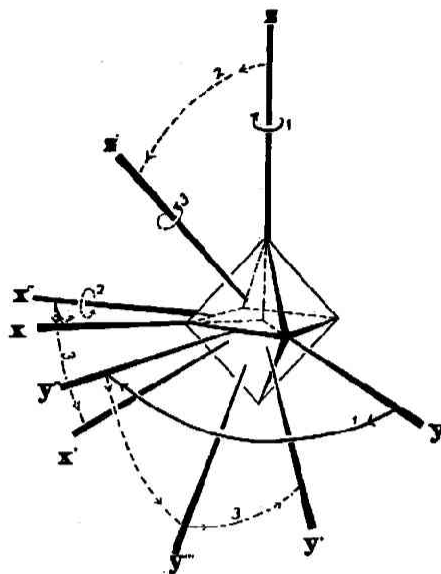


Fig. 3.

$$\left. \begin{aligned} x &= (\cos \alpha \cos \gamma - \sin \alpha \sin \gamma \cos \beta) x' - (\cos \alpha \sin \gamma + \sin \alpha \cos \gamma \cos \beta) y' \\ &\quad + (\sin \alpha \sin \beta) z'. \\ y &= (\sin \alpha \cos \gamma + \cos \alpha \sin \gamma \cos \beta) x' - (\sin \alpha \sin \gamma - \cos \alpha \cos \gamma \cos \beta) y' \\ &\quad - (\cos \alpha \sin \beta) z'. \\ z &= (\sin \alpha \sin \beta) x' + \cos \gamma \sin \beta) y' + (\cos \beta) z'. \end{aligned} \right\} (7)$$

Substituting $\alpha = -45^\circ$, $\beta = -\tan^{-1}\sqrt{2}$ and $\gamma = 45^\circ$ in (7), we obtain

$$\left. \begin{aligned} x &= \frac{\sqrt{3}+1}{2\sqrt{3}} x' - \frac{\sqrt{3}-1}{2\sqrt{3}} y' + \frac{1}{\sqrt{3}} z'. \\ y &= \frac{-\sqrt{3}+1}{2\sqrt{3}} x' + \frac{\sqrt{3}+1}{2\sqrt{3}} y' + \frac{1}{\sqrt{3}} z'. \\ z &= -\frac{1}{\sqrt{3}} x' - \frac{1}{\sqrt{3}} y' + \frac{1}{\sqrt{3}} z'. \end{aligned} \right\} (8)$$

On the other hand, from (1) and (5),

$$\phi_{111} = \sin \theta \cos \varphi + \sin \theta \sin \varphi + \cos \theta.$$

Transforming the polar co-ordinates into the rectangular, we have

$$x^2 + y^2 + z^2 = x + y + z. \quad (9)$$

Substituting (8) in (9), we have

$$x'^2 + y'^2 + z'^2 = \sqrt{3} z'.$$

Transforming again into the polar co-ordinates, we obtain

$$\phi_{111} = \sqrt{3} \cos \theta'. \quad (10)$$

Comparing (1) and (10), we find that ψ_{III} is identical with the original p eigenfunction and the strength is 1.732, considerably weaker than either the tetrahedral or the trigonal eigenfunction. The polar graph of ψ_{III} in the plane containing the z' -axis is given in Fig. 4, and the relation between the functions in (6) is shown in Fig. 5.

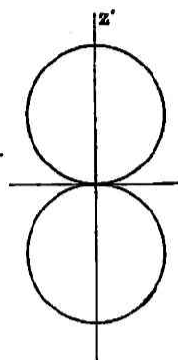


Fig. 4.

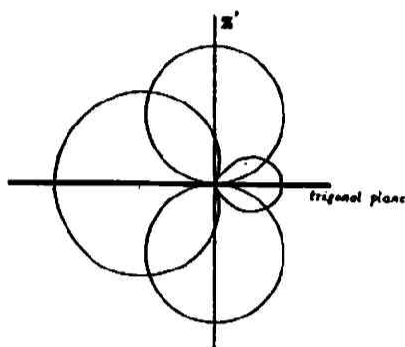


Fig. 5.

For example, the central carbon atom in free triarylmethyl radicals combines with the three aryl radicals by the trigonal functions (4) and the electron is left unpaired in the eigenfunction (5) which is identical with the original p functions.

The benzene molecule is formed by the trigonal eigenfunctions (4), and each carbon atom has a lone electron whose eigenfunction is given by (5) and whose spin is antiparallel to those of the adjacent carbon atoms. The extraordinary stability of the benzene molecule is chiefly due to such lone electrons, the spin of which exercises attraction on both sides of a carbon atom in the ring. It is interesting to note that the benzene rings in diphenyl⁵⁾, terphenyl⁶⁾, quaterphenyl⁷⁾ as well as triphenylbenzene⁸⁾, are all coplanar. These facts may be easily accounted for by the antiparallel spin of the lone electrons of the carbon atoms joining the benzene nuclei. The orbitals (5) of these electrons being perpendicular to the planes of the rings, free rotation about the links between the nuclei is prohibited just as in the ordinary 'double' bond, which will be discussed later. Similarly the benzene rings in the triphenylmethyl radical are expected to lie in a plane. The same must hold for the compounds of condensed rings, such as naphthalene, anthracene, etc. It has been stated in the preceding papers¹⁾³⁾ that the carbon atom in graphite belongs to this category.

5) J. Dahr, *Indian J. Physics*, **7**, 43 (1932).

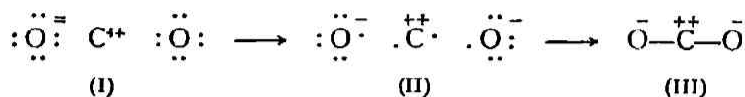
6) L. W. Pickett, *Proc. Roy. Soc., [A]* **142**, 333 (1933).

7) E. Hertel & G. Römer, *Z. physik. Chem., [B]* **23**, 226 (1933).

8) B. Orelkin & K. Lonsdale, *Proc. Roy. Soc., [A]* **144**, 630 (1934).

Digonal Quantization.

In carbon dioxide, two bivalent oxygen anions are co-ordinated around a quadrivalent carbon cation in the following scheme.



As to the behaviour of the bivalent oxygen anion, it has been discussed in the preceeding papers²⁹⁾.

The zeroth order eigenfunctions corresponding to the state (II) above, may be shown as

$$\left. \begin{aligned} \psi_{100} &= \frac{1}{\sqrt{2}} (s + p_x) \\ \psi_{\bar{1}00} &= \frac{1}{\sqrt{2}} (s - p_x) \end{aligned} \right\} \quad (11)$$

These eigenfunctions have maxima in the opposite sense on the x -axis. The maximum value is 1.931, considerably stronger than 1.732 of the original p function (1), but a little weaker than 2.000 of the tetrahedral function (2) and 1.991 of the trigonal quantization (4). The polar graph of the digonal function is shown in Fig. 6.

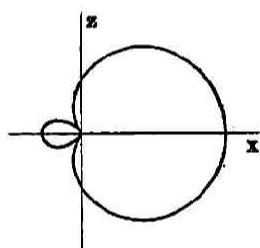
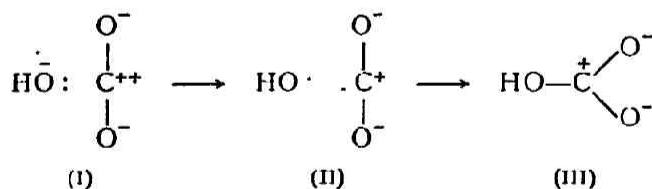


Fig. 6.

Carbon dioxide is readily absorbed in alkaline solutions, whereas it is difficultly soluble in water. This fact may be explained as due to a change in quantization.



Hydroxyl ion is co-ordinated to carbon dioxide molecule in alkaline solutions, changing the digonal quantization into the trigonal. In neutral water, however, the concentration of hydroxyl ion is not sufficient for the change in quantization. Similarly, absorption of sulphur trioxide in alkaline solutions involves a change of the trigonal quantization into the tetrahedral.

Quantization for 'Double' Bond.

The quantization for 'double' bonds is the same as the aromatic quantiza-

tion (6), as has been discussed in the previous paper⁹. The lone electron in the orbital (5), which is identical with the original p eigenfunction and of which the maximum value is given in the direction perpendicular to the trigonal plane of combination, is responsible for all the characteristics of the 'double' bond. In ethylene, for example, the spin of the lone electron in one of the carbon atoms is antiparallel with that in the other carbon atom, and consequently the bond between these two carbon atoms are reinforced, giving rise to the shorter distance than that between two tetrahedral carbon atoms. The instability of the 'double' bond is due to a partial unsaturation of the spin of the lone electrons, e.g., on the outer sides of the carbon atoms in ethylene. Such an unpaired side of an atom may be called an active end, which is open to reactions with other atoms coming into contact. In a conjugate 'double' bond, e.g., in butadiene, the active ends exist only on the carbon atoms (1) and (4), and not on those (2) and (3), the lone electrons on the latter being partially paired on both sides. The anomalous behaviour of the conjugate 'double' bond in addition reaction can thus be explained. Similarly in hexatriene, the active ends are only on the carbon atoms (1) and (6). By linking the carbon atoms (1) and (6) of hexatriene, benzene is obtained, and then the structure is totally devoid of the active ends. The lack of free rotation about the 'double' bond is also due to antiparallel spins of the lone electrons. The planar configuration of ethylene is explained just as in the case of diphenyl.

Quantization for 'Triple' Bond.

By a similar consideration as in the 'double' bond, the quantization for the 'triple' bond has been found.

$$\left. \begin{aligned} \psi_{111} &= \frac{1}{\sqrt{2}}(s + p_z) \\ \psi_{\bar{1}00} &= \frac{1}{\sqrt{2}}(s - p_z) \\ \psi_{010} &= p_y \\ \psi_{001} &= p_z \end{aligned} \right\} \quad (12)$$

It is a combination of the digonal quantization (11) and two of the original p eigenfunctions (1). The former functions give linear configuration and the latter two show the orbitals for two lone electrons. The p orbitals are perpendicular to each other and also to the line of combination of the two atoms. Acetylene is, for example, $\text{H}-\overset{*}{\underset{*}{\text{C}}}-\overset{*}{\underset{*}{\text{C}}}-\text{H}$.

In classical organic chemistry, the number of bonds of a carbon atom has consistently been assumed as four, and consequently the configuration of carbon compounds has been explained by means of the tetrahedral carbon and its modifications. It has, however, been shown that there are carbon atoms with less than four bonds. For examples, in benzene, ethylene, aldehydes, ketones, carboxylic acids, etc. the carbon atoms have three bonds, and in carbon dioxide, acetylene, etc., two. As a matter of fact, the attachment of chemists to the four tetrahedral bonds has provoked many difficulties in dealing with carbon compounds, as has been often pointed out in the previous papers. The present authors would like to emphasize that the valency has nothing to do with the number of bonds, and it is the co-ordination number that decides the latter. This might sound strange to those who are less familiar with complex chemistry. In this branch of chemistry, however, we can find numerous examples of the proposition. For example, platinum in chloroplatinate ion is quadrivalent and has six bonds. As for carbon in question, it is almost always quadrivalent, but the atom may have four, three or two bonds according as the number of ligands it co-ordinates. All the carbon atoms always combine with others by a single bond. There are neither true double bonds nor true triple linkages.

It is quite natural, then, that there are many compounds in which a bond between two carbon atoms is formed by two different kinds of quantization. For example, the side chain of benzyl alcohol is formed by the aromatic quantization on the ring and the tetrahedral function on the chain. One of the most interesting example is the combination between the quantization for the 'double' bond on one side and that for the 'triple' on the other. For example, it is found in an allene derivative, which has been shown as $\begin{matrix} a \\ \diagup \\ C \\ \diagdown \\ b \end{matrix} = C = C \begin{matrix} d \\ \diagup \\ C \\ \diagdown \\ e \end{matrix}$, and is to be represented as $\begin{matrix} a \\ \diagup \\ C^* \\ \diagdown \\ b \end{matrix} - \begin{matrix} * \\ C \\ * \end{matrix} - \begin{matrix} d \\ \diagup \\ C^* \\ \diagdown \\ e \end{matrix}$ in the present system. The central carbon atom is in the quantization for the 'triple' bond and the other two in that for the 'double' bond. Since the two orbitals of the lone electrons in the central carbon is perpendicular to each other, the orbitals of the lone electrons of the outer carbon atoms, each of which is antiparallel with one of those on the central carbon, must naturally be perpendicular to each other. And consequently the plane decided by a, b and C is perpendicular to that by C, d and e. Optically active form can, therefore, result from this configuration.

Summary.

(1) All the carbon compounds are assumed to be formed by co-ordinating ions and molecules around quadrivalent carbon cations.

(2) According to the number and nature of ligands, there are at least six types of bonds in carbon compounds: viz., the tetrahedral, the trigonal, the aromatic, the digonal, the 'double', and the 'triple' bond.

(3) The nature of these valence bonds in carbon compounds have been dealt with from a quantum mechanical point of view.

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